

# **Measurement of Ar-39 in Argon**

Andrew Sonnenschein, David Finley, Stephen Pordes, Richard Schmitt,  
C. J. Martoff

## **ABSTRACT**

Liquid argon is a promising detection medium for WIMP dark matter but the sensitivity of argon detectors is compromised by the presence of the beta-emitting isotope  $^{39}\text{Ar}$  formed in atmospheric argon by cosmic ray interactions. The recent discovery of plentiful sources of underground argon depleted in  $^{39}\text{Ar}$  may enable the construction of multi-ton, background free detectors. Due to limitations in the existing measurement techniques, the full extent of the depletion is unknown and only a few of many potential sources have been characterized. We propose to build a low-background  $^{39}\text{Ar}$  counter capable of measuring  $10^{-3}$  of the normal atmospheric concentration and begin working with other groups to assay possible sources of argon for a large detector at DUSEL. The argon counter will initially be operated in the 100 m deep MINOS near detector tunnel at Fermilab and will be moved to DUSEL as soon as practical.

## 1.1 Introduction

One of the most important motivations for the construction of DUSEL is to provide a site for the operation of large-scale detectors for weakly interacting dark matter particles. Many R&D efforts are underway to understand the possibilities and limitations of the various proposed technologies for these detectors and rapid progress has been made. It seems possible to anticipate that detectors proposed for the initial suite of DUSEL experiments (starting construction around 2012) will have sensitivities three orders of magnitude greater than the currently existing ones, with target masses of a ton or more, and background counting rates below a few events per year [1].

For target masses above 1-ton, the technologies likely to succeed are, in our opinion, (1) Bubble chambers and (2) Noble liquid scintillation chambers, possibly instrumented to collect ionization as well as light. Fermilab has a very active program to address the R&D issues of the bubble chamber technology. The current proposal would begin to involve the laboratory in R&D on noble liquids.

A critical question for the noble liquid detectors is which liquid is the most promising target. The cross section for the scattering of Weakly Interacting Massive Particles (WIMPs) by spin-independent interactions is proportional to the square of the number of nucleons in the target, which for equal masses favors xenon over argon by the ratio  $(131/40)^2 \sim 11$  if energy thresholds are low enough to avoid loss of coherence. On the other hand, argon offers the possibility of dramatically higher levels of background discrimination, due to the very large difference in scintillation pulse shapes for nuclear and electron recoils [2]. Argon is also available in effectively unlimited quantities, and a thousand times less expensive (currently 2 k\$/ton compared to  $\sim 3$  M\$/ton for xenon) which may be an important consideration for large chambers.

When considering standard, commercially available liquid argon as the target liquid, backgrounds are expected due to the beta decay of Ar-39, which is present at the level of  $8 \times 10^{-16}$  of the dominant, stable isotope Ar-40. Commercial liquid argon is a distillation product of liquid air and Ar-39 is introduced into air by neutron interactions on Ar-40 at high altitude. The resulting decay rate is  $\sim 10^5$  /kg-day, with a significant fraction of the counts falling into the 0-100 keV energy window relevant for dark matter searches.

Argon from underground sources, such as natural gas wells and subsurface waters, has the potential to be much lower in Ar-39 than atmospheric argon. Recently, the Princeton group (supported by 2007 DUSEL R&D funds) and collaborators have demonstrated for the first time that at least two underground argon sources are more than ten times lower in Ar-39 than atmospheric argon [3]. They have made a convincing case that extraction of large quantities of underground argon will be feasible and economical. Other groups in the US, Canada and Europe are investigating alternative sources of depleted argon. The

critical questions that must be answered next are “How low in Ar-39 do these sources go?” and “What is the most practical and economical source to exploit for production of multi-ton quantities?”

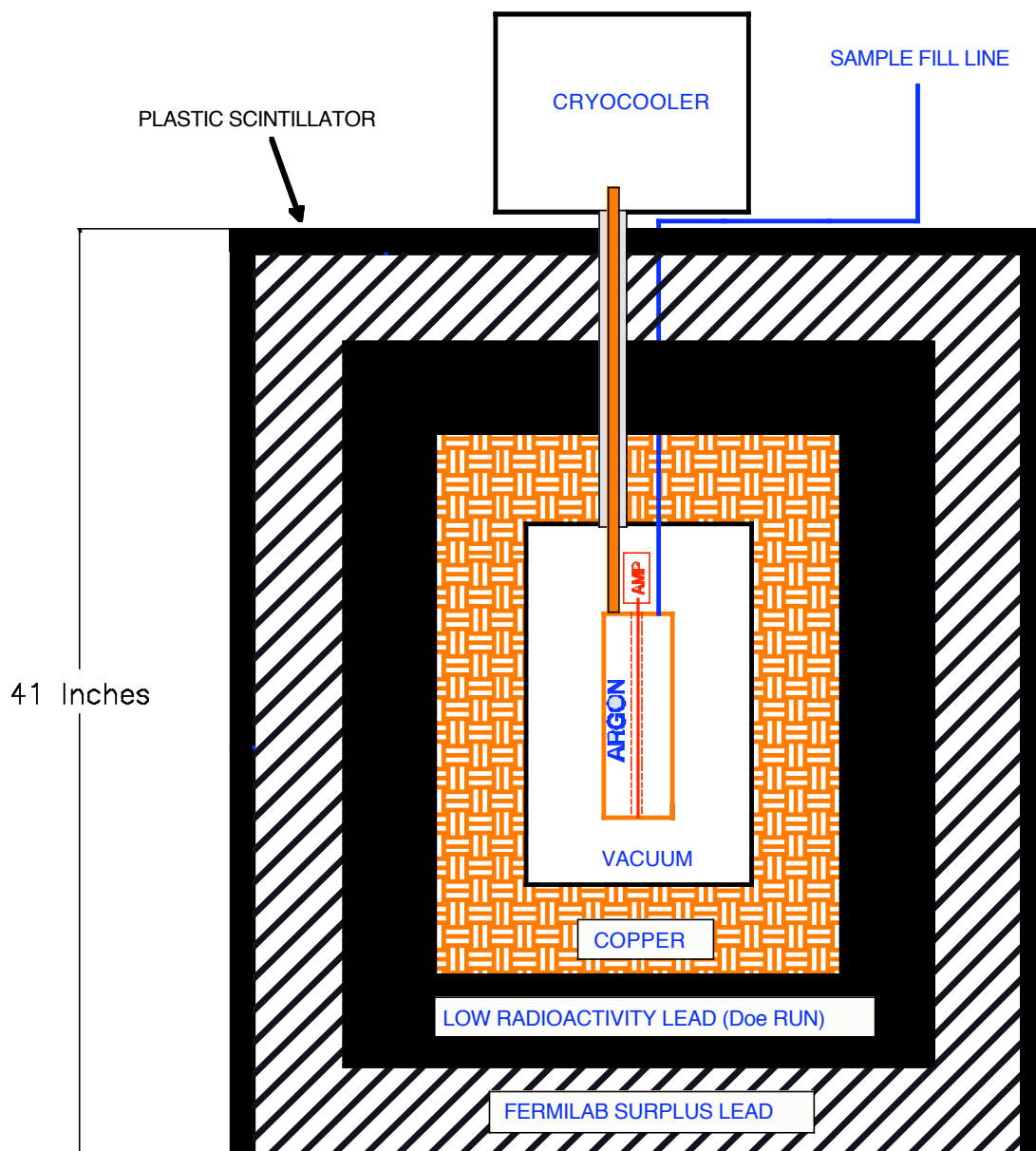
We believe that argon may emerge as the favored noble liquid for dark matter searches if practical sources of sufficiently low-radioactivity argon can be exploited. If Ar-39 is suppressed by three orders of magnitude (or more) with respect to atmospheric argon, simple scaling arguments based on existing proposals suggest that it will be possible to construct ~10-100 ton argon detectors that remain background free for more than a year of running. This would improve sensitivity to WIMPS by more than 4 orders of magnitude beyond the best published limits.

*An essential tool for the development of underground argon sources is a practical counter that can measure the activity of Ar-39 at the relevant levels. We propose to build such a counter and operate it as a screening facility. Given the rate from atmospheric argon of  $10^5$  /kg-day and our intention to be sensitive to  $10^{-3}$  of this rate, our design aims to be able to measure a hundred counts per kilogram per day. The initial site of the screening facility will be the tunnel at Fermilab built for the Minos near detector. When DUSEL has reached an appropriate level of development and on-site operations staff are available, we will transfer the counter to DUSEL. Our partners in counting argon samples will be the Princeton group that has discovered the first useful underground sources. They intend to extract and purify a sample from the National Helium Reserve of sufficient volume to fill our detector within the next 18 months (funding contingent). We have also made contact with a group at Los Alamos that is investigating a different set of sources.*

## **1.2 Design of the argon counter**

Two techniques are presently available for the measurement of Ar-39/Ar-40 isotope ratios; these are low-level counting in small, high-pressure gas proportional counters and accelerator mass spectroscopy [4]. Currently, proportional counters provide the greatest sensitivity, which extends down to about 4% of the atmospheric level ( $3 \times 10^{-17}$ ) if samples of ~ 1 gas liter can be obtained. Accelerator mass spectroscopy currently has a detection limit of about 30% of the atmospheric ratio, but can be applied to samples as small as a few mL.

We propose to construct a low background counting cell for liquid argon, which should have sensitivity to  $10^{-3}$  of the atmospheric ratio. This will be accomplished by counting a relatively large amount of argon (equivalent to 800 gaseous liters) in a cell built with very low background materials and surrounded by a very high quality radiation shield. Our design strategy is to build the simplest possible detector out of the best materials identified for use in recent dark matter and neutrino experiments. The design will allow the counter to be operated with gaseous argon at pressures up to 20 bar. This will give us a capability to measure small gas samples (up to ~100 g) at somewhat reduced sensitivity



**Figure 1: Schematic of the Detector**

in cases where a full liter of liquid argon is not available. This capability may be particularly valuable in the initial stages.

A sketch of our detector and shielding design is shown in Figure 1. The argon will be contained in a cylindrical 1-liter copper vessel, with a single charge sensing wire running down the center. We will drift charge to the central wire and measure it with a low-radioactivity FET preamp attached to the top of the argon vessel. Cooling for the argon will be provided by a copper cold finger extending from an external cryocooler. A small electric heater attached to the cold finger will be used to regulate the argon temperature.

The materials used in the cryostat and detector will be copper, fused silica for insulating structures, plastics for mechanical support; the materials for small fasteners and vacuum seals will be selected carefully. The charge preamplifier will be based on the CDMS cold electronics design. All materials will be screened by low-level counting or ICPMS. We note that our detector is considerably less complex than the germanium detectors which have been used for low background spectroscopy in this energy region. With the appropriate level of attention to radiopurity details, we should be able to achieve equivalent or improved background levels.

### **1.3 *Shielding and expected backgrounds***

Key to the success of the counter is to shield it as well as possible from external radiation that could mimic the Ar-39 signal. The shield design uses the experience from current dark matter experiments and low-level germanium counters. The argon counter will be shielded with 20 cm of lead and 10 cm of copper. The inner 10 cm of lead will come from the Doe Run Company, which operates a mine in Missouri that is known to produce material with relatively low abundance of the Pb-210 isotope, typically  $\sim 20$  Bq/kg, with up to a 50% variation between batches [5]. Past experience with lead shielding for low background experiments has shown that this isotope is usually the only important source of internal activity in virgin lead [6]. The Pb-210 can produce background events through the bremsstrahlung of the betas emitted by its daughter Bi-210 ( $E_{\text{max}}=1.16$  MeV). With no shielding inside the lead, we would expect the background rate from this source near the 200 keV peak in the Ar-39 beta spectrum to be  $\sim 7$  counts/keV-kg-day for Doe Run lead.

The Bi-210 bremsstrahlung is attenuated by about a factor of two by a centimeter of copper shielding [7]. Our shielding design provides sufficient copper to ensure that the contribution of the Bi-210 bremsstrahlung to the background rate will be less than 0.01 events/keV-kg-day. Since this is the only relevant internal activity expected in the lead and the thickness of lead is enough to ensure negligible penetration of external environmental gamma rays, we expect the gamma flux in the shield to be dominated by trace radioisotope contamination of the copper. The purity of the copper will likely determine the ultimate sensitivity of our device. Since many low-radioactivity experiments have used copper, the internal activity of copper has been well studied. Shields using a standard industrial grade of electrolytically-refined copper, known as OFHC (oxygen-free high-conductivity), without extraordinary efforts to avoid the in-growth of cosmogenic activities, typically achieve backgrounds  $\sim 1$  count/keV-kg-day near the Ar-39 beta peak. Recently, an even higher-purity copper has been identified by a group from the Max Planck Institute (MPI) which participates in several experiments at Gran Sasso. They have found a supplier that is willing to work with experimenters to minimize cosmic-ray exposure [8]. This copper has been used to reach background levels of  $\sim 0.07$  counts/keV-kg-day at 200 keV [9]. This is the state of the art in compact low-background gamma shielding, with better results having been obtained only in very large detectors shielded by many meters of high purity water or liquid scintillator. Our

intention is to procure copper of equivalent quality, preferably from a supplier in the United States, to reduce the cosmogenic activity that accumulates during shipping.

Surface contamination could limit the sensitivity of our device if not carefully controlled. Recent CDMS results give an indication of the surface contamination levels that are achievable with good cleaning and handling procedures. The CDMS detectors see a rate of 0.002 beta events per  $\text{cm}^2\text{-day}$  in the region 10-100 keV. Scaling this rate to the  $700 \text{ cm}^2$  interior surface of our detector, we would expect  $\sim 1.4$  events per day background in the 100 keV energy region around the Ar-39 beta peak (0.01 counts/keV-kg-day), which will not limit sensitivity.

Since we intend to initially operate the experiment in the  $\sim 300$  m.w.e. Minos near detector tunnel at Fermilab, where the cosmic muon flux is intense enough to produce counting rates several orders of magnitude larger than our signal through a combination of direct interactions, electromagnetic showers and neutron generation inside the shield, we will surround the detector with a set of plastic scintillator muon veto counters. We expect these counters to have less than a  $10^{-4}$  inefficiency for tagging muons entering the lead shielding, based on the success with similar counters used for other shallow-site experiments (CDMS-I, COUPP). Vetoing counts in the argon within a few hundred microseconds of a passing muon will reduce the muon-induced background to negligible levels.

The MINOS near-detector tunnel also receives a high intensity neutrino pulse from the Fermilab accelerator complex every two seconds, with a duration of about 10 microseconds. Beam-related backgrounds will be removed with a timing cut.

## **1.4 Expected sensitivity**

The Ar-39 isotope is a beta emitter with a 565 keV endpoint and a half-life of 270 years. At the atmospheric abundance of  $8 \times 10^{-16}$  Ar-39/Ar-40, it has a decay rate of 1.4 Bq per liquid liter of argon (1400 grams). The differential decay rate at 200 keV is 300 counts/keV-kg-day. The response of our detector to Ar-39 will be measured very precisely by counting high-activity samples. Given the high counting statistics expected with our device and the well-understood signal spectrum, it should be possible to detect Ar-39 at a signal to background ratio of  $\sim 1:1$ . If we are successful in achieving state-of-the-art background levels ( $< 0.3/\text{keV-kg-day}$ ), we will have sensitivity to  $10^{-3}$  of the atmospheric decay rate.

## 1.5 References

- [1] Dark Matter Working Group White Paper, DUSEL Town Meeting, Washington, DC, Nov. 2-4, 2007.  
[http://cosmology.berkeley.edu/DUSEL/Town\\_meeting\\_DC07/working\\_groups.html](http://cosmology.berkeley.edu/DUSEL/Town_meeting_DC07/working_groups.html)  
(Note: final document to be available at this address in early 2008)
- [2] Boulay, M.G., Hime, A, Astropart. Phys., 25 3 (2006), arXiv:astro-ph/0411358.
- [3] D. Acosta-Kane et al., “Discovery of underground argon with low level of radioactive  $^{39}\text{Ar}$  and possible applications to WIMP dark matter detectors”,  
<http://arxiv.org/abs/0712.0381>
- [4] Collon, P., Kutschera, W., Lu, Z., Annu. Rev. Nucl. Part. Sci, 2004. 54:39-67.
- [5] D.S. Leonard et al., in press, NIM A., arXiv:0709.4524
- [6] G. Heusser, Annu. Rev. Nucl. Part. Sci. 45:643-590.
- [7] A. J. Da Silva, Ph.D. Thesis, University of California, Berkeley, 1996.  
<http://cdms.berkeley.edu/Dissertations/dasilva.pdf>
- [8] H. Neder, G. Heusser and M. Laubenstein, Appl. Rad. Iso, 53 (2000) 191-195. G. Heusser, M. Laubenstein, H. Neder, “Low-level germanium gamma-ray spectrometry at the  $\mu\text{Bq/kg}$  level and the future developments towards higher sensitivity”, preprint.  
[http://www.mpi-hd.mpg.de/ge76/tg11\\_files/GeMPIpub2e4b.ps](http://www.mpi-hd.mpg.de/ge76/tg11_files/GeMPIpub2e4b.ps)
- [9] D. Budjas et al, Proceedings of 14th International Baksan School "Particles and Cosmology-2007", April 16-21, 2007.  
<http://www.mpi-hd.mpg.de/personalhomes/mheisel/Baksan-Heisel.pdf>

